

Kondo destruction and valence fluctuations in an Anderson model

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Unconventional quantum criticality in heavy-fermion systems has been extensively analyzed in terms of a critical destruction of the Kondo effect. Motivated by a recent demonstration of quantum criticality in a mixed-valent heavy-fermion system, β -YbAlB₄, we study a particle-hole-asymmetric Anderson impurity model with a pseudogapped density of states. We demonstrate Kondo destruction at a mixed-valent quantum critical point, where a collapsing Kondo energy scale is accompanied by a singular charge-fluctuation spectrum. Both spin and charge responses scale with energy over temperature (ω/T) and magnetic field over temperature (H/T). Implications for unconventional quantum criticality in mixed-valence heavy fermions are discussed.

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Competing interactions in quantum systems give rise to zero-temperature phase transitions. If it is continuous, such a transition takes place at a quantum critical point (QCP). There is mounting evidence, especially in heavy-fermion systems, that a QCP can underlie unconventional superconductivity [1]; related considerations have been applied to high-temperature cuprate and iron pnictide superconductors [2]. It is standard to describe a QCP within the Ginzburg-Landau-Wilson (GLW) framework: critical destruction of an order parameter characterizing a spontaneously broken symmetry gives rise to collective modes associated with order-parameter fluctuations [3]. In the context of antiferromagnetic metals, this is referred to as a spin-density-wave QCP [4].

Recent experiments in heavy-fermion metals have clearly established the existence of a novel class of antiferromagnetic QCPs, characterized by non-Fermi liquid behavior and ω/T scaling in the dynamical spin susceptibility [5]. There are indications that such unconventional QCPs also promote superconductivity [6]. These QCPs defy description in terms of a GLW functional [7, 8]; their understanding requires the introduction of quantum modes beyond order-parameter fluctuations. The proposed additional modes are associated with the critical destruction of the Kondo effect [7, 8]. In the paramagnetic phase, Kondo singlets form and generate Kondo resonances, thereby turning the local moments into single-electronic excitations and enlarging the Fermi surface. The destruction of the Kondo effect across the antiferromagnetic QCP suppresses the Kondo resonances, making the Fermi surface small. Critical Kondo destruction therefore manifests itself in a discontinuous evolution of the Fermi surface across the transition, as has been observed through quantum oscillation and Hall effect measurements [5, 9].

Theoretical studies of critical Kondo destruction have largely been confined to the Kondo-lattice limit of inte-

ger valence. In rare-earth intermetallics, superconductivity is believed also to arise in the vicinity of valence transitions [10], which have been found to be first order. Until recently, there has been no significant evidence for a QCP associated with valence fluctuations. The situation has changed with the observation of mixed valency in the ytterbium-based heavy-fermion superconductor β -YbAlB₄ [11], which is quantum critical under ambient conditions [12]. In an applied magnetic field, the magnetization obeys H/T scaling [13], consistent with the ω/T scaling seen previously near the unconventional QCPs of antiferromagnetic heavy-fermion compounds. These properties implicate β -YbAlB₄ as a strong candidate for a mixed-valent heavy-fermion QCP, and raise the prospect that the material's unusual scaling behavior can be understood in terms of critical Kondo physics.

At first glance, critical Kondo destruction at mixed valence appears unlikely. Kondo destruction in a Kondo lattice amounts to the localization of f electrons. While unconventional, this is physically transparent, because localization can readily arise for a commensurate filling of an electronic orbital (one f -electron per site). At mixed valence, the situation is more subtle because the f orbital has a fractional, generally incommensurate, per-site occupancy, and there is no mechanism known for electron localization at incommensurate fillings. This leads to important questions of principle: Can critical Kondo destruction occur in the presence of valence fluctuations and, if so, how does the criticality compare to its local-moment counterpart? For instance, are charge excitations part of the critical fluctuation spectrum?

In this Letter, we address these issues in the mixed-valence regime of an Anderson impurity model whose conduction-electron density of states features a pseudogap centered on the Fermi energy. We focus on an impurity model because of the local nature of the Kondo-destruction physics; formally, the Kondo lattice model

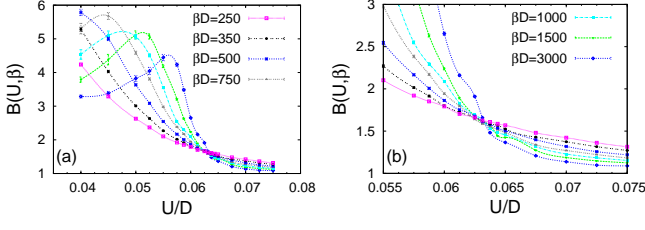


FIG. 1: (a) Binder cumulant $B(U, \beta)$ vs. U for $r = 0.6$, $\Gamma_0 = 0.1D$, and $\epsilon_d = -0.05D$, and the labeled temperatures. Error bars were obtained from a jackknife analysis of the CT-QMC data. (b) Blow up of the same data around the intersection of curves, which determines $U_c/D = 0.063125 \pm 0.0008$.

can be treated through an effective impurity model in the extended dynamical mean field approach [14, 15]. Given that the commensurate-filling (i.e., local-moment) limit of the model exhibits critical Kondo destruction and associated dynamical scaling properties [16], we consider the pseudogapped density of states to provide a prototype setting to search for a Kondo-destruction QCP at mixed valence. Our model has the advantage of being amenable to study using reliable methods: the continuous-time quantum Monte Carlo (CT-QMC) method [17] and the numerical renormalization group (NRG) [16, 18, 19].

Surprisingly, we do find critical Kondo destruction in this mixed-valent model. The critical properties in the spin sector reflect the collapse of an energy scale as the QCP is approached from the Kondo-screened side but not from the Kondo-destroyed side, much as in the integer-valent (local-moment) limit. By contrast, the charge sector shows a collapsing energy scale on both sides of the QCP. The critical point displays H/T (and ω/T) scaling. This existence proof for a Kondo destruction QCP at mixed valence makes it feasible to interpret the H/T and related scaling properties of β -YbAlB₄ in terms of an interacting fixed point. We note that the same model is also relevant to impurity physics in d -wave superconductors and graphene, where the density of bulk fermionic states goes to zero at the chemical potential [20].

The Anderson impurity Hamiltonian is

$$H = \sum_{k,\sigma} [\epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + V (d_\sigma^\dagger c_{k\sigma} + \text{H.c.})] + \epsilon_d n_d + U n_{d\uparrow} n_{d\downarrow}, \quad (1)$$

where $c_{k\sigma}$ annihilates a conduction-band electron of energy ϵ_k , d_σ annihilates an electron of energy ϵ_d in the impurity level, U is the electron-electron repulsion within the impurity level, V is the hybridization taken to be momentum independent, $n_{d\sigma} = d_\sigma^\dagger d_\sigma$, and $n_d = n_{d\uparrow} + n_{d\downarrow}$. The band density of states vanishes in a power-law fashion at the Fermi energy ($\epsilon_F = 0$):

$$\rho(\epsilon) = \sum_k \delta(\epsilon - \epsilon_k) = \rho_0 |\epsilon/D|^r \Theta(D - |\epsilon|). \quad (2)$$

The impurity-band interaction is completely specified by

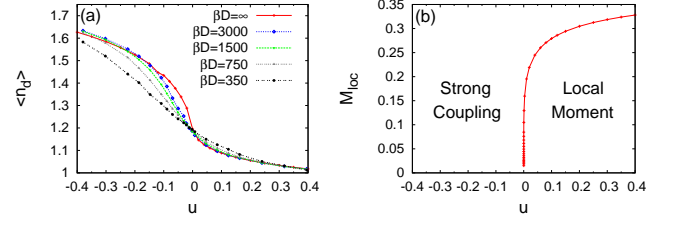


FIG. 2: Valence and local spin properties vs. $u = U/U_c - 1$ for $r = 0.6$, $\Gamma_0 = 0.1D$, and $\epsilon_d = -0.05D$: (a) Occupancy $\langle n_d \rangle$ at the labeled temperatures [25]. The QCP ($u = 0$) occurs at mixed valence, i.e., $\langle n_d \rangle \neq 1$. (b) Local magnetization M_{loc} , showing quenching of the impurity spin for $u < 0$ but the emergence of a free local moment for $u > 0$.

the imaginary part of the hybridization function, $\Gamma(\epsilon) = \pi \sum_k V^2 \delta(\epsilon - \epsilon_k) = \Gamma_0 |\epsilon/D|^r$, where $\Gamma_0 = \pi \rho_0 V^2$.

The critical properties of the model with particle-hole (p-h) symmetry ($\epsilon_d = -U/2$) and its Kondo limit ($U \gg \Gamma_0$, where local charge fluctuations are negligible) have been investigated in a number of analytic and numerical studies [16, 19, 21–23]. The breaking of p-h symmetry is irrelevant for pseudogap exponents r in the range $0 < r < r^* \simeq 0.375$, but becomes relevant for $r > r^*$, leading to a mixed-valent QCP [19]; $r = 1$ serves as an upper critical “dimension”, above which the critical properties have a mean-field character [16, 22].

Here, we investigate the p-h-asymmetric pseudogap Anderson model by varying U for fixed Γ_0 and ϵ_d to pass from a Kondo-screened strong-coupling phase ($U < U_c$) to a Kondo-destroyed local-moment phase ($U > U_c$). We apply the CT-QMC technique, which was recently shown to be able to reach temperatures T sufficiently low to access the quantum critical regime [23]. We measure the dynamical local spin and charge susceptibilities, $\chi_s(\tau, \beta) = \langle T_\tau S_z(\tau) S_z(0) \rangle$ and $\chi_c(\tau, \beta) = \langle T_\tau : n(\tau) : : n(0) : \rangle$, respectively, where $S_z = \frac{1}{2}(n_\uparrow - n_\downarrow)$, $:n: = \sum_\sigma n_\sigma - \langle \sum_\sigma n_\sigma \rangle$, and $\beta = 1/T$ (taking $k_B \equiv 1$) plays the role of the system size. The corresponding static susceptibilities follow from $\chi_{c,s}(\beta) = \int_0^\beta d\tau \chi_{c,s}(\tau)$. Measuring powers of the local magnetization $\langle M_z^n \rangle = \langle [\frac{1}{\beta} \int_0^\beta d\tau S_z(\tau)]^n \rangle$ allows construction of the Binder cumulant [24] $B(U, \beta) = \langle M_z^4 \rangle / \langle M_z^2 \rangle^2$. We supplement our $T > 0$ (finite- β) CT-QMC results with static quantities calculated arbitrarily close to $T = 0$ ($\beta = \infty$) using the NRG method as adapted to treat pseudogap impurity problems [16, 18, 19]. NRG results presented below were obtained with Wilson discretization parameter $\Lambda = 9$, with Γ_0 corrected [19] to compensate for the band discretization, and retaining all many-body states up to 50 times the effective bandwidth of each iteration.

We focus our discussion on the representative case of a pseudogap exponent $r = 0.6$ with $\Gamma_0 = 0.1D$ and $\epsilon_d = -0.05D$. Figure 1 plots the variation of the Binder cumulant with U at different temperatures. For small

U , charge fluctuations are strong and the Binder cumulant lies above the range $1 < B(U, \beta) < 3$ obeyed by a pure spin system [23, 24]; in this limit, the low-energy behavior is close to that of a pseudogap resonant level. As U increases, charge fluctuations are suppressed leading at low temperatures to $B(U, \beta) < 3$; this part of the strong-coupling phase exhibits a true pseudogap Kondo effect. For very large U , by contrast, $B(U, \beta)$ tends towards 1 at low temperatures, suggesting the presence of a decoupled impurity spin, characteristic of the local-moment phase. We locate the phase boundary by the intersection of $B(U, \beta)$ curves for different temperatures [23] at $U_c/D = 0.06313 \pm 0.0008$. The NRG gives $U_c = 0.06450D$, a small shift that can likely be attributed to residual effects of NRG discretization. The mixed-valent nature of the QCP is demonstrated in Fig. 2(a), where the local occupation $\langle n_d \rangle$ is seen to differ from unity at $U = U_c$. Note also that $\langle n_d \rangle$ displays significant temperature dependence in the vicinity of the QCP.

We are now in a position to look for a critical destruction of the Kondo effect in this mixed-valent QCP, i.e., the continuous vanishing of an effective Kondo energy scale signaled by the divergence of the zero-temperature static local spin susceptibility χ_s as U approaches U_c from below. Such a divergence is indeed seen in our zero-temperature χ_s vs. U data [Fig. 3(a)] and in the temperature dependence of χ_s at $U = U_c$ [Fig. 3(b)]. Figure 2(b) shows the U dependence of the local magnetization $M_{\text{loc}} = \lim_{h \rightarrow 0} \lim_{T \rightarrow 0} \langle M_z \rangle$, where h is a local magnetic field entering a term $h(n_\uparrow - n_\downarrow)/2$ (with $g\mu_B \equiv 1$) added to Eq. (1). Since $M_{\text{loc}} = 0$ throughout the strong-coupling phase, and M_{loc} rises continuously from zero on entry to the local-moment phase, this quantity serves as an order parameter for the quantum phase transition. Our results can be summarized as

$$\begin{aligned} \chi_s(T, U = U_c) &\sim T^{-x_s}, \\ \chi_s(T = 0, U < U_c) &\sim |u|^{-\gamma_s}, \\ M_{\text{loc}}(T = 0, U > U_c) &\sim u^{\beta_s}, \end{aligned} \quad (3)$$

where $u = U/U_c - 1$. We find $x_s = 0.80(3)$ from CT-QMC, in excellent agreement with the NRG value $x_s = 0.7908(3)$; the NRG also yields $\gamma_s = 1.42(2)$ and $\beta_s = 0.1874(2)$. These power-law behaviors are all defining characteristics of critical Kondo destruction.

To probe valence fluctuations near the QCP, we turn to the static local charge susceptibility $\chi_c(T, U)$. As shown in Fig. 4(a), $\chi_c(T = 0, U)$ increases with U in the strong-coupling phase and diverges as $U \rightarrow U_c^-$, in a manner similar to $\chi_s(T = 0, U)$. In the local-moment phase, the spin and charge responses are very different: $\chi_s(T = 0, U) = \infty$ but $\chi_c(T = 0, U)$ remains finite, although it diverges as $U \rightarrow U_c^+$. In other words, the valence fluctuation energy scale is nonzero in both phases, vanishing only when U approaches U_c from either side. At $U = U_c$, χ_c has a singular temperature dependence as

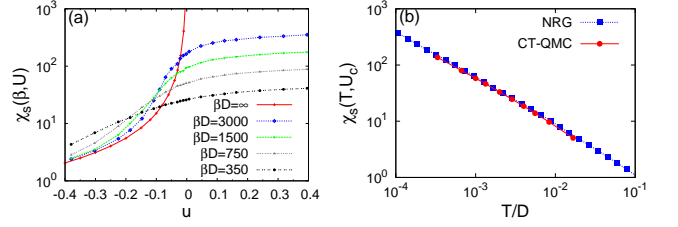


FIG. 3: Local static spin susceptibility $\chi_s(T, U)$ for $r = 0.6$, $\Gamma_0 = 0.1D$, and $\varepsilon_d = -0.05D$, (a) vs. $u = U/U_c - 1$ at the labeled temperatures [25], and (b) vs. T at the critical point $U = U_c$.

shown in Fig. 4(b). These behaviors are consistent with

$$\begin{aligned} \chi_c(T, U = U_c) &\sim T^{-x_c}, \\ \chi_c(T = 0, U) &\sim |u|^{-\gamma_c}. \end{aligned} \quad (4)$$

CT-QMC yields $x_c = 0.36(3)$, while the NRG gives $x_c = 0.120(1)$ (extracted at temperatures much lower than can be accessed by CT-QMC) and $\gamma_c = 0.21(1)$. The difference between the two x_c values stems from a very slow crossover to the quantum critical regime [Fig. 4(b) inset]. The much wider crossover window for χ_c compared with χ_s [Fig. 3(b)] likely arises because $x_c < x_s$, meaning that lower temperatures must be reached before sub-leading contributions to χ_c become negligible. We stress that the singularity of the charge susceptibility is unique to the mixed-valence QCP, and does not appear at its integer-valence counterpart.

We reach the important conclusions that the Kondo destruction occurs at a genuinely mixed-valent QCP and that valence fluctuations are part of the critical spectrum. Calculations for level energies $\varepsilon_d \neq -0.05D$ (and hence different critical occupancies $\langle n_d \rangle$) indicate that the critical exponents defined above depend on the band exponent r , but not on the impurity valence. This implies that the divergence of the static charge susceptibility is a universal property. At the same time, we find that the critical behavior in the spin sector coincides with the model in its integer valence limit, i.e., the p-h-asymmetric pseudogap Kondo model [16].

We now discuss the dynamical scaling of $\chi_s(\tau, T)$ and $\chi_c(\tau, T)$. In analogy with the spin response at the Kondo destruction QCP in the usual Kondo limit [23, 26], we find that at $U = U_c$ both $\chi_s(\tau, T)$ and $\chi_c(\tau, T)$ collapse onto the conformal scaling form, showing a power-law dependence on $\pi T / \sin(\pi \tau T)$ with exponents $\eta_{c,s}$. For the temperatures considered, the charge susceptibility has not yet reached its asymptotic power-law behavior [based on Fig. 4(b)]. Our results thus imply that both leading and sub-leading terms of the critical $\chi_c(\tau, T)$ scale in terms of $\pi T / \sin(\pi \tau T)$. The scaling form means $\chi_s(\omega, T)$ and $\chi_c(\omega, T)$ obey ω/T scaling [23] at $U = U_c$.

We next consider the effect on the QCP of applying a finite local magnetic field h . Consistent with the ω/T

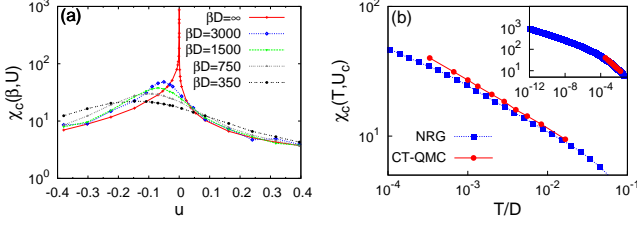


FIG. 4: Local static charge susceptibility $\chi_c(T, U)$ for $r = 0.6$, $\Gamma_0 = 0.1D$, and $\varepsilon_d = -0.05D$, (a) vs. $u = U/U_c - 1$ at the labeled temperatures [25], and (b) vs. T at the critical point $U = U_c$, where the discrepancy between CT-QMC and NRG data is due mainly to the difference in U_c values. Inset: $\chi_c(T, U_c)$ over a wider range of T , showing the slow crossover behavior.

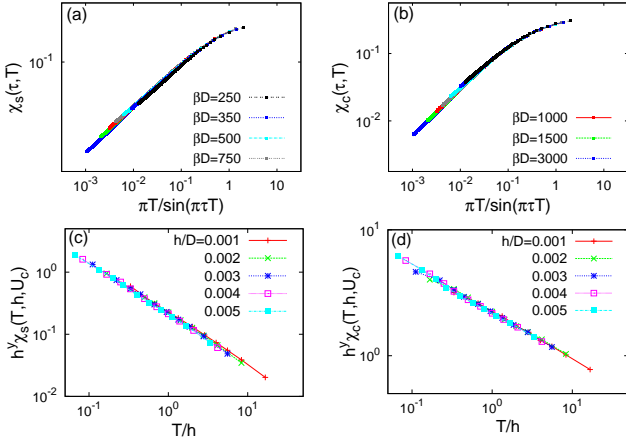


FIG. 5: Scaling of critical spin and charge responses for $r = 0.6$, $\Gamma_0 = 0.1D$, $\varepsilon_d = -0.05D$ and $U \simeq U_c$: (a,b) Dynamical spin and charge susceptibilities vs. $\pi T / \sin(\pi \tau T)$. Both susceptibilities show excellent scaling collapse over two decades of $\pi T / \sin(\pi \tau T)$, with $\eta_s = 0.20(3)$ and $\eta_c = 0.67(3)$. (c,d) Static spin and charge susceptibilities vs. T/h in the labeled local magnetic fields h .

scaling we find T/h scaling for fields $|h| < T_K$, i.e.,

$$\chi_{c,s}(T, h, U_c) \sim h^{-y_{c,s}} f_{c,s}(T/h), \quad (5)$$

where $f_{c,s}(x)$ is a scaling function [see Figs. 5(c) and 5(d)]. Scaling collapse in T/h further reflects the interacting nature of this mixed-valence QCP. At such an interacting QCP, critical exponents satisfy hyperscaling relations that imply $y_{c,s} = x_{c,s}$, equalities confirmed by our results to within numerical accuracy.

Our results are to be contrasted with the generalization of the spin-density-wave QCP to the valence sector [27, 28]. Like its spin counterpart [4], such a mixed-valent QCP is Gaussian (noninteracting) and is not expected to obey either energy-over-temperature or field-over-temperature scaling.

We now briefly consider the case $r = 1$, motivated by critical Kondo screening in graphene and d-wave su-

perconductors. For $\Gamma_0 = 0.1D$ and $\varepsilon_d = -0.05D$, we find $U_c/D = 0.05475 \pm 0.0006$ with CT-QMC, or 0.05562 with NRG. Both methods indicate that the local static charge susceptibility diverges at U_c , along with the spin susceptibility. We find x_c to be an increasing function of r , but logarithmic corrections to scaling [16] prevent reliable determination of critical exponents for $r = 1$.

This work provides new insights into the unusual critical properties of β -YbAlB₄ [11], suggesting that Kondo destruction can occur in this material even though it is mixed valent. (Mixed valence is natural in this material given that its onset Kondo temperature is high—on the order of 200 K—and its mass enhancement is moderate.) The demonstration of h/T scaling provides evidence that the experimentally observed field-over-temperature scaling signals Kondo destruction. Our finding of a rapid variation of $\langle n_d \rangle$ near the QCP, which can be tested experimentally [29], suggests that the concentrated lattice system is essentially quantum critical over a range of densities, leading to the exciting possibility of quantum criticality occurring over a region of parameter space rather than just at an isolated point. Finally, our work raises intriguing questions about the extent to which quantum-critical magnetic and valence degrees of freedom influence the superconductivity observed in β -YbAlB₄.

From a general theoretical perspective, how quantum criticality can go beyond the GLW framework of order-parameter fluctuations is a fundamental problem that is important not only for heavy-fermion metals but also for QCPs arising in insulating magnets and other strongly correlated systems. At present, there are few concrete theoretical examples for such unconventional QCPs. By identifying a new QCP in this category, our results provide another setting to gain intuition about beyond-GLW QCPs in general.

In summary, we have shown that mixed-valent quantum criticality can display the phenomenon of Kondo destruction. The quantum critical point has a collapsing Kondo energy scale and a singular charge-fluctuation spectrum. The valence varies strongly with temperature near the critical point. In the concentrated lattice case, similar quantum critical behavior is expected to occur over an extended range of parameters. Our results raise the prospect of unconventional quantum criticality in mixed-valent systems beyond β -YbAlB₄.

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